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Report:

Introduction

In contrast to conventional Mössbauer spectroscopy performed in the energy domain, the nuclear resonant forward scattering (NFS) of synchrotron radiation is successfully employed as a time-differential technique ([1, 2] and citations therein). The phased excitation of an ensemble of nuclei gives rise to effects like speed up and dynamical *beats* due to the thickness of the sample. The hyperfine interaction of the coherently excited nuclei exhibits a quantum beat structure, i. e. an additional modulation of the time behaviour of the nuclear decay.

The high photon flux of a new 3rd generation synchrotron radiation source like ESRF allows beside NFS, where coherent processes are involved, also nuclear inelastic scattering (NIS) experiments. NIS proceeds incoherently with energy transfer from and to the heat bath made up by the phonons of the system.

Experimental Setup

The measurements were performed at the Nuclear Resonance Beamline ID 18 [3].

$Fe(tpa)(NCS)_2$

We complemented the studies on the polycristalline spin-crossover complex $Fe(tpa)(NCS)_2$ [4,5] (20 % enriched in ^{57}Fe , "tpa" = tris(2-pyridylmethyl)amine) by performing NFS measurements at different temperatures. The hexacoordinate iron(II) shows a temperature induced spin-state transition from the paramagnetic high-spin ($^5T_2$) phase to the diamagnetic low-spin ($^1A_1$) phase.

Evaluation of the spectra with CONUSS

The calculations of NFS spectra have been carried out using the CONUSS program package [6]. The measurements yield a temperature dependence of the fraction of molecules in the high-spin state which is in agreement with the data previously obtained using SQUID. The Lamb-Mössbauer factor shows a dramatic decrease with increasing temperature.

^{57}Fe(OOH)

Superparamagnetism is characteristic for single-domain magnetic particles. At sufficiently low temperatures the magnetization of such particles appears to be static on the Mössbauer time scale, while at elevated temperatures (in the intermediate region) it begins to flip by 180” back and forth at a rate which is comparable with nuclear Larmor frequencies giving rise to interesting dynamic features. Finally, the flipping rate becomes so high that the magnetic hyperfine field cancels to zero and only the remaining electric quadrupole interaction is observable.

A decrease of the forward scattering intensity in the intermediate region is observed in the superparamagnetic systems ^{57}FeOOH and Bacterioferritin from Streptomyces olivaceus as in the paramagnetic system $[^{57}Fe^{II}(CH_3COO(TP_{piv}P)]^-$ [7]. In order to investigate the vanishing forward scattering intensity in the intermediate region in more detail we measured also the nuclear inelastic scattering intensity of nanosized ^{57}Fe(OOH) particles. We found that the intensity in the NIS channel continuously decreases with increasing temperature up to 80K; in this temperature range the intensity of the NFS channel goes through a minimum around 40 K. This behaviour indicates that NFS reflects the destruction of coherence due to the spin dynamics while NIS does not (probably because of experimental conditions).

Evaluation of the spectra with SYNFOS

The dynamics of a flipping spin can be appropriately treated by a stochastic model. This treatment has been implemented recently in a new version of the SYNFOS program package [8,9]. The detailed evaluation of the time spectra is in progress.

Acknowledgments

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